

Electrodynamics of superconducting pnictide superlattices

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It has been recently reported (S. Lee *et al.*, Nature Materials **12**, 392, 2013) that superlattices where layers of the 8% Co-doped BaFe₂As₂ superconducting pnictide are intercalated with non superconducting ultrathin layers of either SrTiO₃ or of oxygen-rich BaFe₂As₂, can be used to control flux pinning, thereby increasing critical fields and currents, without significantly affecting the critical temperature of the pristine superconducting material. However, little is known about the electron properties of these systems. Here we investigate the electrodynamics of these superconducting pnictide superlattices in the normal and superconducting state by using infrared reflectivity, from THz to visible range. We find that multi-gap structure of these superlattices is preserved, whereas some significant changes are observed in their electronic structure with respect to those of the original pnictide. Our results suggest that possible attempts to further increase the flux pinning may lead to a breakdown of the pnictide superconducting properties.

Iron-based superconductors, as well as high temperature cuprate superconductors, are intrinsically multilayered materials. Particular efforts have thus been devoted to the deposition of thin superconducting films and to artificially synthesize heterostructures based onto different superconducting materials. The study of these systems can indeed help in understanding the mechanism of superconductivity itself and can offer the possibility of tailoring important superconducting properties. In particular, high-field applications, which probably represent today the most important technological advantage offered by novel superconductors, require extremely high values of critical current J_c and upper critical field H_c . While, till now, the enhancement of these quantities has been studied in multilayers mainly based on the high- T_c cuprate YBaCuO,^{1,2} only recently epitaxial growth techniques provided artificially engineered superlattices (SL) based on the BaFe₂As₂ (Ba-122) pnictide, which enable strong vortex pinning and can thus produce very large J_c and H_c enhancements.³ In particular, it has been shown that intercalating the Co-doped Ba-122 pnictide with non superconducting layers of either SrTiO₃ (STO-SL) or oxygen-rich undoped Ba-122 (O-Ba122-SL), substantially enhances the critical currents, especially for magnetic fields parallel to the *ab*-plane. On the other hand, the critical temperature does not vary significantly, ranging from 19.7 K in STO-SL, to 21.9 K in the pristine Co-doped film, up to 22.6 K in O-Ba122-SL.³

In spite of their interest, little is known about the electronic properties of these superlattice (SL) films. In particular, it is unclear to what extent the multilayered

structure affects the multiband character of the Ba-122 superconductor.^{4,5} Understanding this point is crucial for the possibility of further enhancing the critical fields in pnictide systems, without modifying significantly the superconducting properties of the pristine material.

We employ here infrared spectroscopy to study the electronic structure of pnictide SLs in the normal and superconducting states. Infrared spectroscopy is indeed a powerful technique to investigate both charge dynamics and band structure as it probes both free carriers and interband excitations and can provide a separation of the different contributions to the frequency dependent conductivity.^{6,7} Furthermore, measurements performed in the THz region can provide direct information about presence and nature of superconducting energy gaps.^{7,8} In the case of the Ba-122 system, several infrared studies have been performed, in particular in the THz region (for a recent review, see Ref. [9])

The three samples under investigation are a 400 nm thick film of 8% Co-doped Ba122 (Co-Ba122-film), and the STO-SL and O-Ba122-SL introduced above. Both STO-SL and O-Ba122-SL are made of 24 repetitions of a 13 nm Co-doped Ba122 layer, intercalated with either 1.3 nm STO or 3 nm O-Ba122 layer. All samples are grown on a substrate, made of a SrTiO₃ (STO) template (40 nm thick) deposited on (La,Sr)(Al,Ti)O₃ (LSAT). More details on sample preparation and morphology can be found in Ref. [3]. For comparison THz measurements were also performed on a film of undoped Ba-122.

The temperature dependent absolute reflectivities $R(\omega)$ in the normal state (25-300 K) are measured with

the help of a BRUKER 70v interferometer. By using gold and aluminum mirrors as reference and various beam-splitters, detectors and thermal sources, $R(\omega)$ spectra extend over a frequency range from THz to visible. To measure the reflectivity in the superconducting state we use THz synchrotron radiation from the SISSI beamline¹⁰ of the Elettra synchrotron. The reflectivity ratio R_S/R_N (i.e. the ratio between the 6 K superconducting and the 25 K normal state reflectivity) is measured by cycling the temperature above and below T_c , without moving the sample. The measure of the reflectance ratio, as well as that of the transmittance ratio, has been introduced long ago (see the seminal work of Ref. [11] and references therein), and is still widely used today in the study of different superconducting systems.^{9,12–14} Indeed, by exploiting synchrotron radiation, this technique can provide high accuracy results intrinsically unaffected by possible misalignments between sample and reference.

The reflectivities of the three samples measured at 300 and 25 K are reported in Fig. 1a up to 8000 cm^{-1} . In the inset, the $R(\omega)$ spectrum of the Co-Ba122-film is shown in all the explored spectral range. For all samples, the effect of temperature is only evident below 3000 cm^{-1} , where the low temperature reflectivities are slightly enhanced with respect to room temperature, as a typical consequence of a decreased scattering rate. The $R(\omega)$ of the Co-Ba122-film displays a pronounced edge at about 2000 cm^{-1} , typically attributed to the multiband nature of this material. The $R(\omega)$ of the STO-SL is very similar to that of the film, though slightly depressed. On the other hand the $R(\omega)$ of the O-Ba122-SL has a different shape, with a less pronounced bend at 2000 cm^{-1} and a higher level in the mid- and near-infrared range.

To describe the complex conductivity of the system, which determines its optical response,^{6,7} we use a two-band model. While density functional theory¹⁵ predicts up to 5 bands crossing the Fermi level, it has been proven that simplified models in which only two bands are considered, capture the essential low-energy physics in iron-based superconductors.^{16,17} The model includes the sum of two Drude terms (parallel conductivity model¹⁸) and of Lorentz terms accounting for possible interband transitions. This two-band Drude-Lorentz model has been widely employed in describing the optical response of Ba122 system (see for example Ref. [9] and refs. therein). In the present case, the fitting procedure¹⁹ takes into account, besides the finite thickness of the samples, the possible reflections from the substrate via Fresnel equations.⁷ This procedure requires the knowledge of the optical properties of the substrate, which were obtained from *ad-hoc* measurements on the STO+LSAT substrate by using a standard Lorentz-model fitting.^{6,7}

The $R(\omega)$ spectra can be fitted by using, besides the two Drude terms, a first broad Lorentz term mimicking electronic interband transitions in both mid- and near-infrared, and a second one accounting for contributions coming from the ultraviolet.^{20,21} An example of the best fit curve is reported in the inset of Fig. 1. The optical conductivity $\sigma_1(\omega)$ is recalculated according to the model

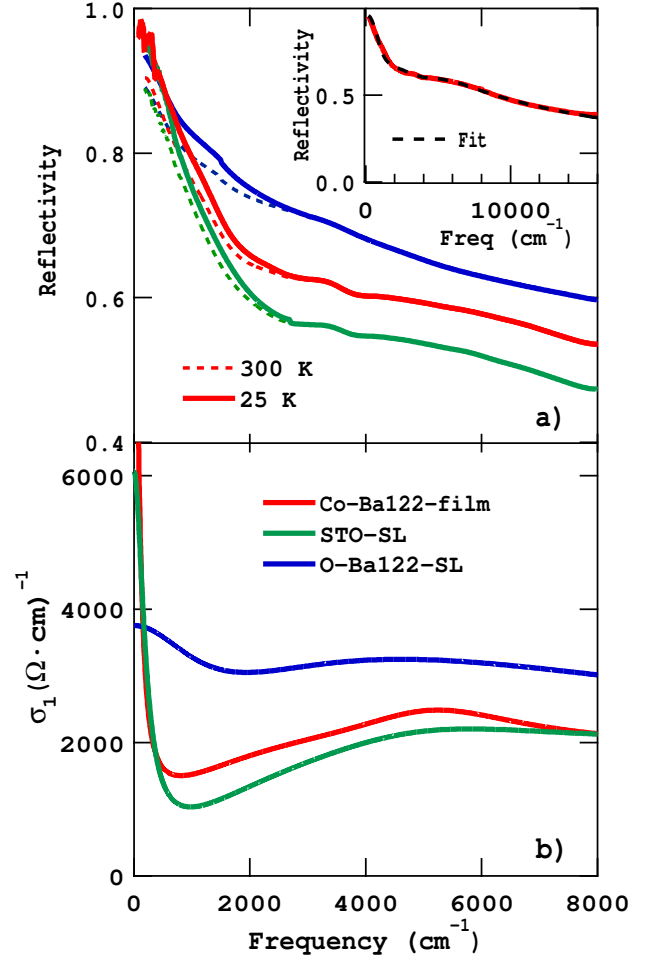


FIG. 1. Color online. a) Optical Reflectivity $R(\omega)$ of the three samples (Co-Ba122-film, STO-SL, and O-Ba122-SL) at 25 and 300 K. Inset: Full range $R(\omega)$ of the Co-doped film, and corresponding best fit curve (see text). b) Optical conductivity of the three samples at 25 K, as recalculated from the two-band Drude-Lorentz model.

and displayed in Fig. 1b. A clear separation between the two Drude bands is well distinguishable for both the Co-Ba122-film and the STO-SL, since the scattering rates of these two Drude bands are about $100\div 200 \text{ cm}^{-1}$ (narrow Drude) and 4000 cm^{-1} (broad Drude). We recall that the presence of two Drude bands with very different widths has been adopted by many authors to describe the optical properties of $\text{BaFe}_{2-x}\text{Co}_x\text{As}_2$ systems (see for example Refs. [20–22]). The broad peak centered at 5000 cm^{-1} can be associated to an electronic interband transition.²¹ In the O-Ba122-SL case, the situation is much different. The first Drude term becomes strongly damped and less discernible from the electronic background due to the presence of the second broad Drude component. A broadening of the peak at 5000 cm^{-1} is also observed.

The increased scattering rate observed in the normal state of the O-Ba122-SL seems to be at odds with the better crystalline quality of this compound with respect to

STO-SL, as revealed by x-rays. As reported in Ref. [3], the crystalline quality is influenced by the better structural matching of the O-Ba122 layers on the Co-Ba122 matrix. To reconcile this scenario with our observations, we note that not only the narrow Drude band scattering rate increases, but also the overall optical conductivity is strongly enhanced in the O-Ba122-SL. This should be interpreted as an increase in the number of charge carriers probably due to the oxygen enrichment, acting like a dopant. Moreover, the broadening of the peak at 5000 cm^{-1} also indicates non-negligible effects on the band structure. Qualitatively, we may think that while the band structure of STO and Co-Ba122 are fully disconnected thus creating energetic barriers between the various portions of the sample, the situation with O-Ba122 is much more different. In fact one may expect the bands to be continuously connected (band bending), with a consequent modification of both band structure and chemical potential in the proximity of the interface. The presence of the O-Ba122 intercalation thus seems to affect the electronic structure in a more profound way, beyond the possible issues related to crystallinity and disorder.

One may thus wonder whether multiband superconductivity is preserved at all in the superlattice compounds and in particular in O-Ba122-SL. To answer this question we turn to the THz electrodynamics in the superconducting state. We compare in Fig. 2a the measured reflectivity ratio R_S/R_N for the Co-Ba122-film, together with that of the undoped, non superconducting, Ba122 film. As expected,^{7,8} the R_S/R_N spectrum clearly deviates from a straight line, as a consequence of superconducting gap(s) opening in the Co-Ba122-film case, while it is flat and very close to 1 in the case of the non-superconducting Ba122 film. It is worth noting that the R_S/R_N spectral shape is very similar to those reported in previous works on different Co-doped Ba122 films.^{9,23}

To describe the electrodynamics of the superconducting systems here investigated, the same procedure employed in the analysis of the THz response of different two-band systems (such as Co-doped Ba122 films,^{9,23} MgB_2 ,¹⁸ and V_3Si ¹²) has been used. In this procedure, each Drude term is substituted by the Zimmermann term,²⁴ which describes the electrodynamics of a superconductor of arbitrary purity, thus generalizing the standard BCS Mattis-Bardeen model.^{7,8} By employing the same Drude-Lorentz normal state parameters obtained from the fittings of Fig.1, we obtain superconducting gap values $\Delta_A=18$ and $\Delta_B=60\text{ cm}^{-1}$, for the narrow and broad Drude terms, respectively.

In the case of the STO-SL (Fig.2b) and O-Ba122-SL (Fig.2c), R_S/R_N has a shape which is reminding that of the Co-Ba122-film. The main difference is in the onset of the R_S/R_N enhancement, which is found at about 40 cm^{-1} , slightly lower than in the Co-Ba122-film ($\approx 60\text{ cm}^{-1}$). The shape of R_S/R_N at the lowest frequencies (i.e. below 25 cm^{-1}), is more flat in the Co-Ba122-film, with respect to the two SLs. The fitting results¹⁹ (see Figs. 2b and c) show that, in both cases, the smaller

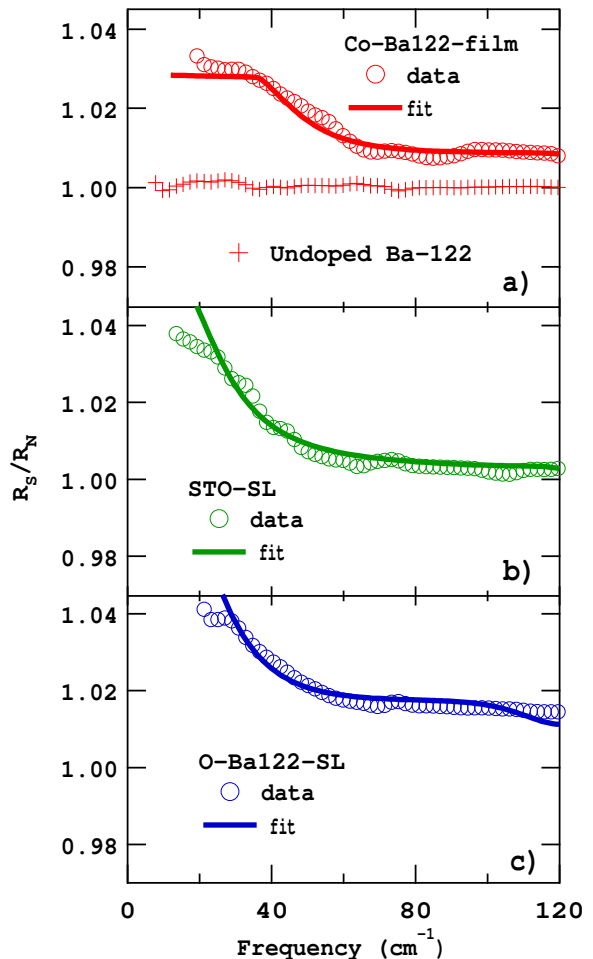


FIG. 2. Color online. a) Reflectivity ratio $R_S/R_N = R(6K)/R(25K)$ for an undoped and a 8%Co-doped Ba122 film (Co-Ba122-film). The full red line is the best fit curve of the Co-Ba122-film data (see text). b) Reflectivity ratio and best fit curve for the STO-SL. c) Same as before for the O-Ba122-SL.

gap Δ_A reduces down to about 7 cm^{-1} , while Δ_B essentially remains unchanged.

It is clear that the inclusion in the model of an additional third electronic channel, ungapped or characterized by a very small gap, may further increase the fit quality, especially on the low frequency side. However, the present model already captures the essential physics we are interested in. In particular, the main message is that the multi-gap structure in the superconducting state is preserved in both SLs. This result is far from trivial in the light of the important differences observed in the normal state properties, especially for what concerns the O-Ba122-SL. The main difference between the Co-doped Ba122 film and the two SLs is the decrease of the smaller gap value Δ_A . Interestingly, such a decrease does not affect significantly neither the larger gap Δ_B nor T_C .

We have addressed here the electrodynamic properties of the pnictide superlattices STO-SL and O-Ba122-SL.

The infrared data show that the normal state electronic properties of the STO-SL are very similar to those of the Co-Ba122-film. On the contrary, the O-Ba122-SL has different electronic properties with, in particular, a less pronounced multi-band structure. However, we have verified that both SL systems remain multi-gap superconductors, even though the small gap value Δ_A is slightly reduced with respect to the value observed in the Co-Ba122-film. In conclusion, our results point out that, albeit the multi-gap structure is still preserved in both SLs, one should be aware of the risks related to changes occurring in the electronic properties while attempting to further increase pinning centers, especially in the case of the oxygen enrichment technique.

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